

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE



In re application of:

Paul EUSTACE et al.

Art. Unit: 1712

Appl. No: 10/049,604

Examiner: BUTTNER, DAVID J.

Confirmation No: 9719

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Atty. Docket No: 31229-178457

For: MELT-PROCESSABLE  
THERMOPLASTIC  
COMPOSITIONS

Customer No:  
  
26694

PATENT TRADEMARK OFFICE

**DECLARATION UNDER 37 C.F.R. §1.132**

Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

Sir:

I, Nicholas Marston, a named inventor of US patent application 10/049,604 and a Senior Research Scientist of Lucite International UK Limited of Wilton Centre, Wilton, Redcar, TS10 4RF, United Kingdom, have derived, or under my supervision there was derived, the impact resistance and the surface gloss of the following melt processable compositions prepared as outlined in US patent application 10/049,604.

**Example 1** Preparation of cross-linked acrylic particulate copolymers having different amounts of alkyl acrylate comonomer

The cross-linked acrylic copolymers (Examples 1a to 1e as detailed in Table 1) were made by bulk polymerisation in a sealed bag with a monomer mixture comprising the specified amounts of methyl methacrylate (MMA), n-butyl acrylate (nBA), ethylene glycol dimethacrylate (EGDMA – cross-linking monomer), dodecyl mercaptan (DDM – a chain transfer agent), LL55 (a UV absorber) and lauryl peroxide (a polymerisation initiator). The mixture was subjected to a pre-programmed thermal polymerisation cycle and then cooled. The resultant polymer was ground and sieved through a standard US 60 mesh sieve. The sieved fractions of Examples 1a to 1e were employed in Example 2.

Table 1: Cross-linked acrylic copolymers with varying amounts of n-butyl acrylate comonomers.

<i>Example no.</i>	<i>MMA % by wt</i>	<i>nBA % by wt</i>	<i>EDGMA % by wt</i>	<i>DDM % by wt</i>	<i>LL55 % by wt</i>	<i>Lauryl peroxide % by wt</i>
1a	77.85	20	0.6	0.5	1	0.05
1b	82.85	15	0.6	0.5	1	0.05
1c	87.85	10	0.6	0.5	1	0.05
1d	89.95	8	0.6	0.5	1	0.05
1e	92.85	5	0.6	0.5	1	0.05

**Example 2**

Each of the cross-linked acrylic copolymers of Examples 1a to 1e (8% by weight) were compounded with 92% by wt of an acrylic polymer of methyl methacrylate and ethyl acrylate and pelleted using a Werner Pfleiderer twin screw extruder (230 °C, 275 rpm). The resulting pellets were co-extruded to form 1mm thick and 100mm wide sheet samples 2a to 2e respectively.

The gloss values of each sheet sample was derived according to ASTM D3679 at a 75° observation angle. The results are presented in Table 2 and Figure 1.

Table 2: Gloss Values

<i>Sheet</i>	<i>75° Gloss</i>
2a	62
2b	59
2c	57
2d	56
2e	55

Typically the gloss value of an acceptable sheet should be less than 75. It is clear from Table 2 and Figure 1 that all sheet samples 2a to 2e exhibit an acceptable gloss value.

The impact resistance of each of the sheet samples was derived in accordance with ASTM D4226. The results are presented in Table 3 and Figure 2.

Table 3: Impact Resistance

<i>Sheet</i>	<i>Impact Resistance (in-lb)</i>
2a	87
2b	80
2c	72
2d	69
2e	65

Typically, the mean failure height of an acceptable sheet is 65 in-lb. It is clear from Figure 2 that all sheet samples 2a to 2e exhibit an acceptable impact resistance. However, sheet 2e formed from a particulate copolymer comprising 5% by wt n-butyl acrylate comonomer represents the minimum amount of comonomer present in the particulate copolymer in order to produce a sheet having the desired impact resistance.

Example 3 Preparation of cross-linked acrylic particulate copolymers having different amounts of cross-linker.

The cross-linked acrylic copolymers (Examples 3a to 3e as detailed in Table 4 were prepared as described in Example 1).

Table 4: Cross-linked acrylic copolymers with varying amounts of cross-linker.

<i>Example no.</i>	<i>MMA % by wt</i>	<i>nBA % by wt</i>	<i>EDGMA % by wt</i>	<i>DDM % by wt</i>	<i>LL55 % by wt</i>	<i>Lauryl peroxide % by wt</i>
4a	82.35	15	1.1	0.5	1	0.05
4b	82.85	15	0.6	0.5	1	0.05
4c	83.05	15	0.4	0.5	1	0.05
4d	83.15	15	0.3	0.5	1	0.05
4e	83.35	15	0.1	0.5	1	0.05

Each of the cross-linked acrylic copolymers of Examples 3a to 3e were compounded with an acrylic polymer of methyl methacrylate and ethyl acrylate and formed into sheets 4a to 4e, respectively, as detailed in Example 2.

The gloss values of each sheet sample 4a to 4e was derived according to ASTM D3679 at a 75° observation angle. The results are presented in Table 5 and Figure 3.

Table 5: Gloss Values

<i>Sheet</i>	<i>75° Gloss</i>
5a	34
5b	59
5c	70
5d	75
5e	85

As mentioned in Example 2 above the gloss value of an acceptable sheet is less than 75. It is clear from Table 5 and Figure 3 that sheets formed from a cross-linked acrylic particulate copolymer having less than 0.3% by weight cross-linker (sample 5e) produce a sheet having an unacceptable gloss level.

The impact resistance of each of the sheet samples 4a to 4e was derived in accordance with ASTM D4226. The results are present in Table 6 and Figure 4.

Table 6: Impact Resistance

Sheet	Impact Resistance (in-lb)
4a	69
4b	80
4c	84
4d	86
4e	91

It is clear from Figure 4 that all of the sheets 4a to 4e have the desired impact resistance of greater than 65 in-lb. However, sheet 4a formed from a particulate cross-linked acrylic polymer including 1.1% by wt of cross-linker has an impact resistance of 69 in-lb which is close to the acceptable lower limit of 65 in-lb.

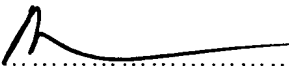
#### Conclusion

The impact resistance and the gloss value of a sheet produced in accordance with US patent application no. 10/049,604 depends on both the amount of acrylic comonomer and the amount of cross-linker in the particulate acrylic copolymer. In particular, particulate copolymers having 5 to 20% by wt of acrylic comonomer and 0.3 to 1.1% by wt cross-linking comonomer produce sheets having the desired impact resistance and gloss value.

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All statements made of my own knowledge are true and all statements made on information and belief are believed to be true. I acknowledge that willful false statements and the like are punishable by fine or imprisonment, or both, under 18 U.S.C. 1001.

Nicholas Marston:



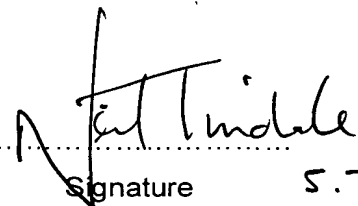
Date:

06.07.2004

Witnessed by:

NEIL TINDALE

Name



Signature

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Figure 1

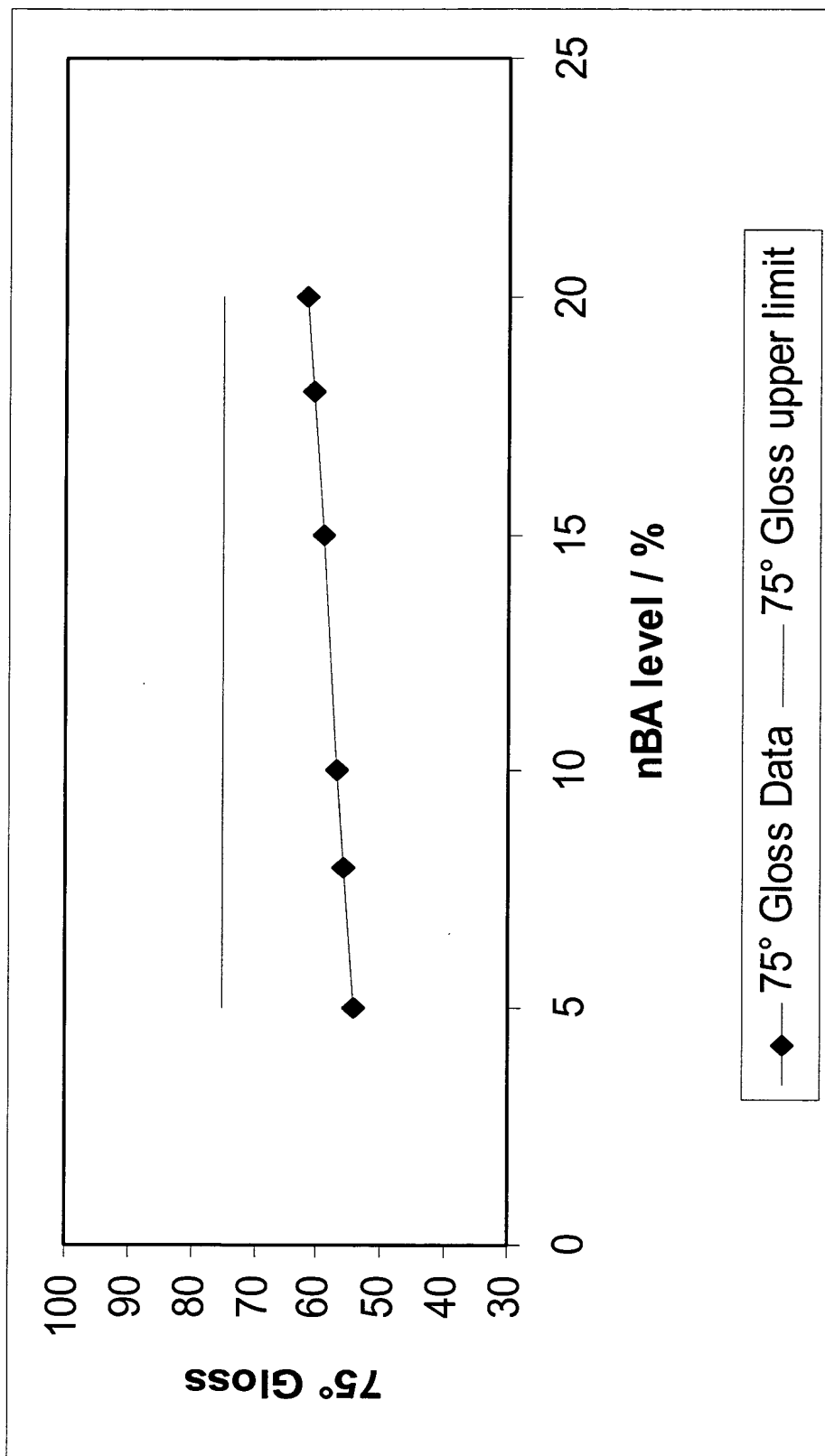


Figure 2

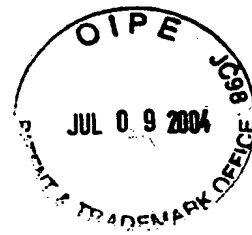
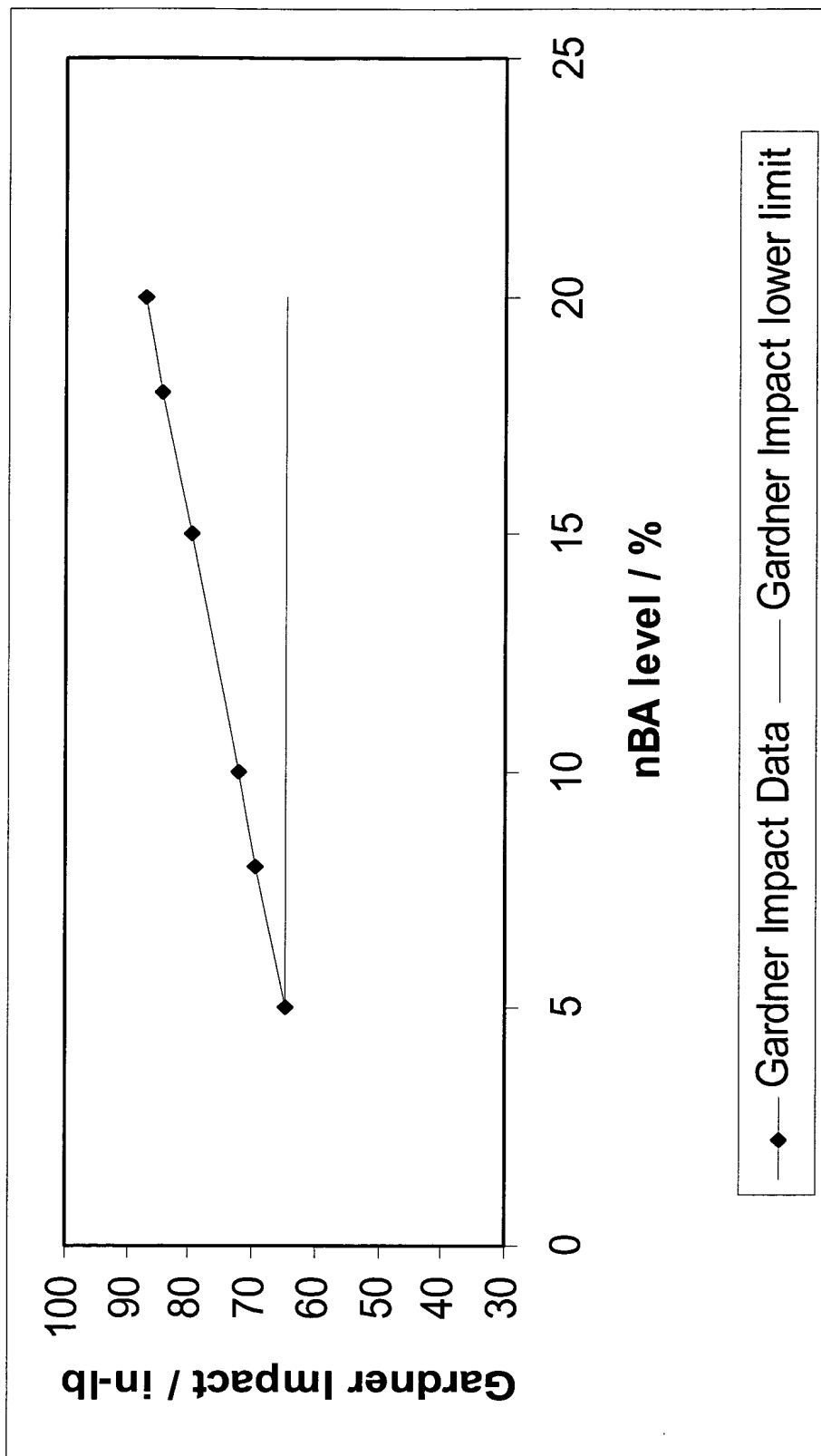


Figure 3

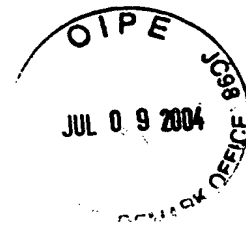
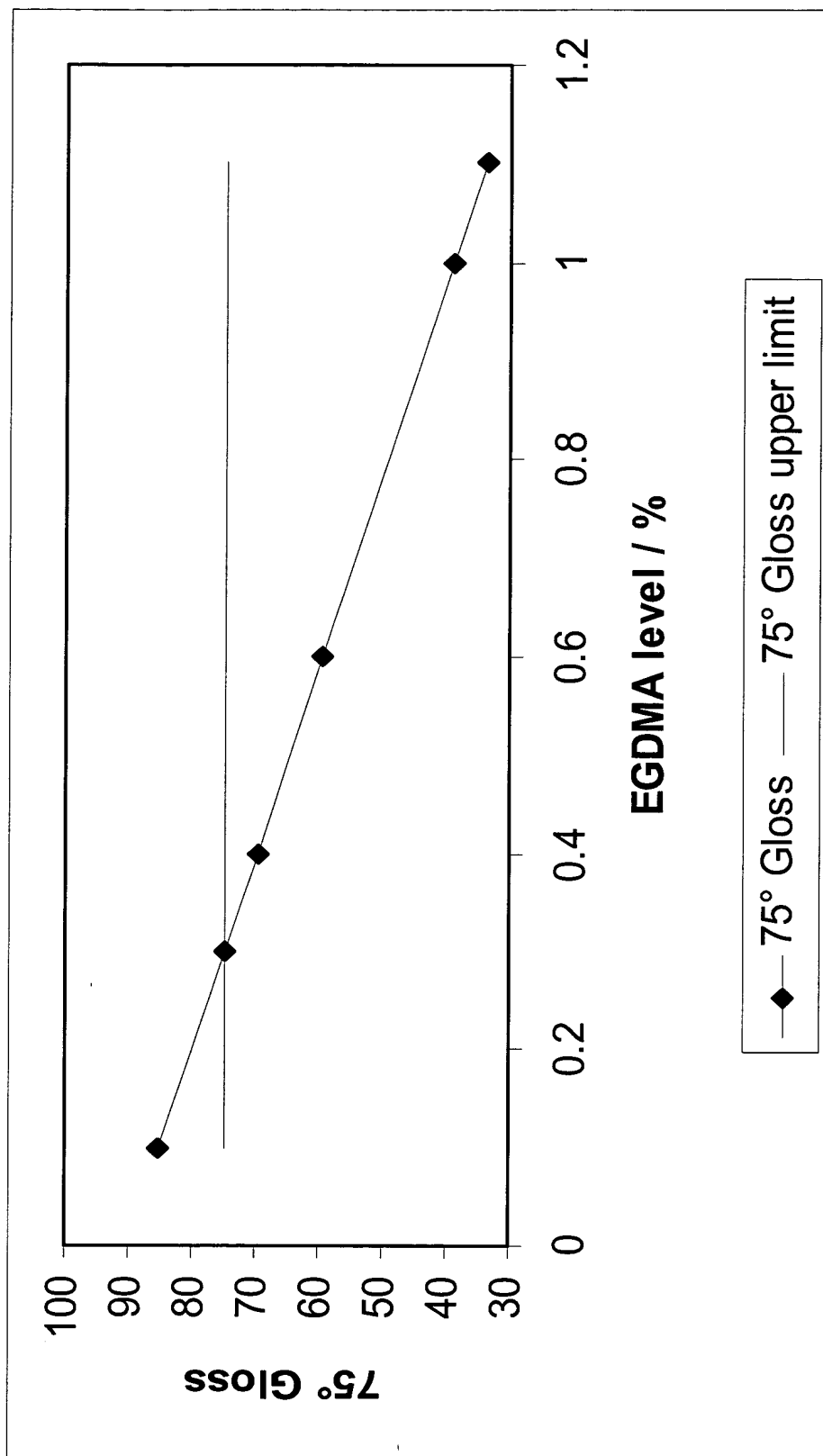




Figure 4

